Magnetization plateaus as insulator-superfluid transitions in quantum spin systems

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We study the magnetization process in two-dimensional $S=1/2$ spin systems, to discuss the appearance of a plateau structure. The following three cases are considered: (1) the Heisenberg antiferromagnet and multiple-spin exchange model on the triangular lattice, (2) the Shastry-Sutherland-type lattice [which is a possible model for $SrCu_2(BO_3)_2$], (3) the 1/5-depleted lattice (for CaV_4O_9). We find in these systems that magnetization plateaus can appear owing to a transition from superfluid to a Mott insulator of magnetic excitations. The plateau states have charge-density-wave order of the excitations. The magnetizations of the plateaus depend on components of the magnetic excitations, the range of the repulsive interaction, and the geometry of the lattice.

In some one-dimensional spin systems, spin-density-wave states with finite spin gap appear under a finite magnetic field accompanying plateau structures in the magnetization process. Magnetization plateaus were observed in some quasi-one-dimensional materials.[1] Theoretical arguments clarify that the appearance of the plateau is explained by an insulator-conductor transition of magnetic excitations.[2] In two- or higher-dimensional systems, magnetization plateaus have been also found in both theoretical[3–5] and experimental studies.[6–8] In this paper, we propose a rather general picture that these two-dimensional plateaus are formed owing to field-induced insulator-superfluid transitions of magnetic excitations. To demonstrate how it works, we discuss three examples in details.

The first example is a family of antiferromagnets on a triangular lattice. For the $S=1/2$ antiferromagnet on a triangular lattice (AFT), Nishimori and Miyashita[3] found a magnetization plateau at $m/m_{\text{sat}}=1/3$, which comes from the appearance of a collinear state with three sublattices, i.e., the so-called ''uud'' state. This plateau was actually observed in AFT materials like C_6Eu (Ref. 6) and $CsCuCl_3$ (Ref. 7). Recently in a multiple-spin exchange (MSE) model, which is a possible model[9] for solid ^{3}He films, a magnetization plateau was predicted[5] at $m/m_{\text{sat}}=1/2$. In this case, the plateau is attributed to the formation of a similar collinear state but with four sublattices. The magnetization processes of these systems have been studied extensively and here we just attempt *interpreting* the known results to test the new picture.

We take as the second example the $S=1/2$ Heisenberg antiferromagnet (HAF) on the Shastry-Sutherland lattice (Shastry-Sutherland model, hereafter, see Fig. 1),[10] which is known to have an exact dimer ground state. Recently Kageyama *et al.*[8] found that $SrCu_2(BO_3)_2$ realizes a lattice structure equivalent to that discussed in Ref. 10 and that it has a gapful ground state. The magnetization measurements show plateaus at $m/m_{\text{sat}}=1/8$ and 1/4. The last is the $S=1/2$ HAF on the 1/5-depleted square lattice (Fig. 2), which includes a model Hamiltonian for CaV_4O_9. In this system, the plaquette singlet state is realized in the ground state.[11]

In our picture, the plateau states can be regarded as Mott insulators of *effective* magnetic particles; repulsive interactions induce various kinds of charge-density-wave (CDW) long-range order leading to a finite energy gap in particle-hole excitations. Except for the plateau phases, magnetic particles are conducting to form supersolids, in which superfluidity and CDW coexist, and magnetization increases smoothly. Of course, the charge density is translated into the spin (S^z) density and superfluidity here means long-range order in the direction perpendicular to the field. Although this essential picture is common to the three examples, the concrete forms of the magnetic particles are different.

In the first example, i.e., AFT and an MSE model, a single flipped spin itself works as the magnetic particle, while the triplets on the dimer (J) bonds are the relevant particles of the Shastry-Sutherland model. We find plateaus at $m/m_{\text{sat}}=1/2$ and 1/3. In the third one, i.e., the 1/5-depleted square lattice, plaquette triplets behave as particles. We predict plateaus at $m/m_{\text{sat}}=1/8$, 1/4, 1/2.

The magnetizations where a plateau appears depend on (i) the form of the magnetic excitations, (ii) range of the repulsion between them, and (iii) the geometry of the lattice. Finally, we summarize common features on properties of the phase transition.

Spin = magnetic particle. The magnetization plateau for the AFT system was found in spin 1/2 anisotropic[3] and isotropic[4] Heisenberg models. The Hamiltonian is

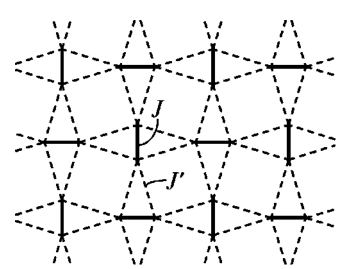

FIG. 1. Shastry-Sutherland lattice.

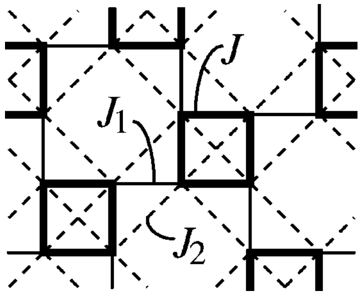

FIG. 2. 1/5-depleted square lattice.

$$H=J\sum_{\langle i,j\rangle}(S_i^xS_j^x+S_i^yS_j^y+\eta S_i^zS_j^z)-B\sum_i S_i^z, \qquad (1)$$

where the summation runs over all nearest-neighbor pairs and B denotes the magnetic field. For $\eta\geqslant 1$, the magnetization curve has a plateau at $m/m_{\mathrm{sat}}=1/3$. The ground state in the plateau phase is of a collinear structure with three sublattices, where two of three spins direct upward and the other downward. In the other phases, magnetic states have noncollinear structures with off-diagonal long-range order (ODLRO). If we introduce a particle picture, i.e., recognize spin dynamics as induced by the motion of a certain kind of particles,[12] the appearance of plateau is easily understood from simple consideration about compressibility of the particles. Regarding an up spin as a hard-core boson and a down one as a vacancy,[12] we can rewrite the Hamiltonian (1) as

$$H=\frac{J}{2}\sum_{\langle i,j\rangle}\{(b_i^\dagger b_j+\mathrm{H.c.})+2\eta n_in_j\}-(B+3J\eta)\sum_i n_i, \qquad (2)$$

where $b_i^\dagger$ denotes the creation operator of the hard-core boson on site i, and n_i is the number operator. Since particles carry magnetic moment unity, the chemical potential $\mu=B+3J\eta$ is controlled by the magnetic field and the μ dependence of the particle density n corresponds to the magnetization curve of the original spin system.

The hopping term comes from the spin-exchange (XY) term and the repulsive interaction from the diagonal (Ising) part. The anisotropic case, $\eta>1$, is mapped to the strong coupling (i.e., strong repulsion) region of the corresponding boson system. The particle-hole transformation converts the system into that of holes with repulsion of the same strength; in the strong-coupling limit, the ground state at the filling $n=2/3$ ($m/m_{\mathrm{sat}}=1/3$) has the density-wave long-range order, with the three-sublattice structure shown in Fig. 3(a). Due to the repulsive interaction, this state is incompressible, i.e., $dn/d\mu=0$, and density-fluctuation energy has a finite gap above the ground state. Except for the filling(s) $n=2/3$ (and 1/3), there are vacancies and hence particles are mobile (conducting). Since the particles obey boson statistics and the system is uniform, the system presumably shows superfluidity. There is perfect correspondence between the above consideration and the previous results;[3,4] the insulating CDW state with $dn/d\mu=0$ is consistent with the spin collinear state, where susceptibility is vanishing. On the other hand, superfluidity of bosons corresponds to noncollinear ODLRO of spins.

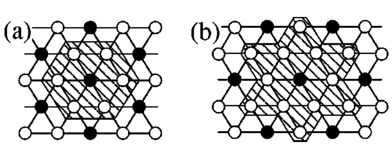

FIG. 3. Spin-density-wave order in AFT (a) and MSE model (b). Black (white) circles denote down (up) spins.

The particle density where CDW stabilizes depends on the range of repulsive interactions. To see this, we next discuss the MSE model with four-spin exchange on the triangular lattice, where repulsion acts further than in the Heisenberg model. The Hamiltonian is given by

$$\mathcal{H}=J\sum_{\langle i,j\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j+K\sum_p h_p-B\sum_i \sigma_i^z, \qquad (3)$$

where $\boldsymbol{\sigma}_i$ denote Pauli matrices. The second summation runs over all minimum diamond clusters and h_p is the four-spin exchange $h_p=4(P_4+P_4^{-1})-1$ with the ring permutation of four spins P_4. It was shown that three- and four-spin exchange interactions are very strong in two-dimensional solid ^{3}He due to strong quantum fluctuations.[9] Theoretically a magnetization plateau was found[5] at $m/m_{\mathrm{sat}}=1/2$ instead of $m/m_{\mathrm{sat}}=1/3$. In the particle picture, bosons feel the following two-body repulsion:

$$V=4(J+5K)\sum_{\langle i,j\rangle}n_in_j+4K\sum_{\substack{(i,j)\\ \in \mathrm{N.N.N.}}}n_in_j. \qquad (4)$$

The repulsive interaction acts in both nearest- and next-nearest-neighbor sites. Figure 3(b) shows the region where the interaction works. Because of the range of repulsion, the particles can solidify at the density $n=1/4$. (Note that the solidification occurs only if the repulsion overcomes the effect of the hopping term.) This insulating phase at $n=1/4$ corresponds to the magnetization plateau at $m/m_{\mathrm{sat}}=1/2$ in the original spin system. The previous numerical result in Ref. 5 on the ground state of the plateau phase is consistent with the CDW order shown in Fig. 3(b).

Dimer triplet. When specific pairs of two spins are more strongly coupled than to the others by an antiferromagnetic interaction, the dimer singlet state realizes in the ground state. Under a weak magnetic field, $S^z=1$ triplets on the dimer bonds are dominant excitations. Several types of repulsive interactions between these dimer triplet excitations can induce various insulating phases and thereby yield magnetization plateaus.

For concreteness, we discuss the Shastry-Sutherland model[10] shown in Fig. 1. The exact dimer ground state[10] is realized for $J'/J<0.69$. (Ref. 13). Recently Kageyama *et al.*[8] found this lattice structure in $SrCu_2(BO_3)_2$ and observed the magnetization plateaus at $m/m_{\mathrm{sat}}=1/8$ and 1/4. Because of the special structure of the lattice, a triplet excitation is almost localized.[13] Considering the dimer triplet state with $S^z=1$ as a particle (a hard-core boson by definition) and the dimer singlet as a vacancy, we derive an effective Hamiltonian for it using the perturbational expansion from the $J'=0$ limit. The expansion is performed up to the third order in J'/J from degenerate states with a constant number of dimer triplets. The effective Hamiltonian up to the second order is

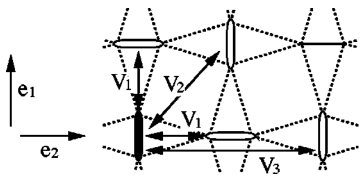

FIG. 4. Two-body repulsive interactions up to the third order of J'/J. $V_1=J'/2+J'^2/2J-J'^3/4J^2$, $V_2=J'^3/8J^2$, $V_3=J'^2/2J+3J'^3/4J^2$.

$$H=\left(J-B-\frac{J'^2}{J}\right)\sum_i n_i+\left(\frac{J'}{2}+\frac{J'^2}{2J}\right)\sum_{\langle i,j\rangle} n_i n_j$$
$$+\frac{J'^2}{4J}\sum_{i\in A}\{[b_i^\dagger(b_{i+e1}-b_{i-e1})+\mathrm{H.c.}](n_{i-e2}-n_{i+e2})$$
$$+2n_{i+e2}(1-n_i)n_{i-e2}+(b_{i+e1}^\dagger b_{i-e1}+\mathrm{H.c.})n_i\}$$
$$+\frac{J'^2}{4J}\sum_{i\in B}\{e1\leftrightarrow e2\}, \tag{5}$$

where $i(j)$ runs over an effective square lattice consisting of dimer bonds (both horizontal and vertical) and horizontal (vertical) ones belong to the $A(B)$ sublattice. The full form of the effective Hamiltonian up to the third order will be published elsewhere.[14] The derived Hamiltonian does not have the one-particle hopping term (as was already reported in Ref. 13), but contains many correlated-hopping processes, where an effective hopping of a particle is mediated by another one. This is one of our main observations. Most third-order terms concern the correlated hopping. Longer-range repulsions between particles appear from higher-order perturbations. Diagonal repulsive interactions up to the third order in J'/J are shown graphically in Fig. 4. The resulting Hamiltonian does not have 90° rotational invariance, since the lattice structure has low symmetry, and this may lead to highly anisotropic CDW states.

We study the effective Hamiltonian in the classical limit. To this end, we map the hardcore boson system to the $S=1/2$ quantum spin system and then approximate the spin-1/2 by a classical vector. We search for the ground state with large sublattice structures (e.g., a stripelike one with six sublattice) both with the mean-field approximation and a Monte Carlo method by decreasing temperatures gradually. The evaluated magnetization process is shown in Fig. 5. Note a clear difference between the high- and low-field region. There appear plateau structures at $m/m_{\mathrm{sat}}=1/2$ and 1/3. The plateau states have CDW long-range orders shown in Fig. 6. Configurations realized for $m/m_{\mathrm{sat}}=1/2$ and 1/3 correspond to perfect closed packings provided that particles avoid repulsion from first- and second-order perturbation, respectively. The plateau at $m/m_{\mathrm{sat}}=1/2$ appears only in the region $0<J'/J<0.50$ and, for large J'/J the CDW is destroyed by the correlated hoppings, which are dominant in the higher-order terms. The correlated hoppings are so efficient also at large particle density that any plateau does not appear for $1/2<m/m_{\mathrm{sat}}<1$. Below $m/m_{\mathrm{sat}}=1/3$, the correlated hoppings occur rarely, because of a low particle density. The observed 1/4 plateau (and 1/8 plateau) of $SrCu_2(BO_3)_2$ may be formed by weak longer-range repulsions which are not taken into account in the present study. Recently Miyahara and Ueda discussed semiphenomenologically that the 1/4 plateau might come from a CDW state with a stripe structure.[15] In our approach, the repulsive interaction relevant to the stripe CDW may come from the higher-order terms in perturbation, otherwise from other spin interactions that are not considered in the Shastry-Sutherland model. This remains a future problem.

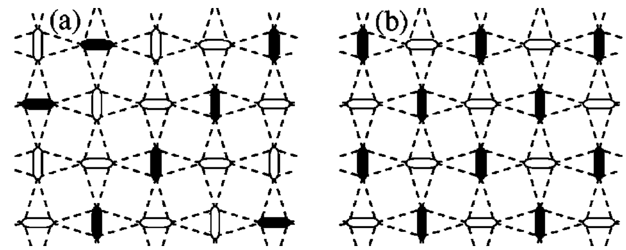

FIG. 6. Spin configurations at magnetization plateaus at $m/m_{\mathrm{sat}}=1/3$ (a), 1/2 (b). Black bonds denote dimer triplet excitations.

Plaquette triplet. When the interactions or a special geometry of the lattice allows four-spin plaquettes, in each of which four spins are coupled more strongly than to the others, individual plaquettes form singlets in the ground state. The triplet states with $S^z=1$ on plaquettes are dominant excitations in a weak magnetic field. The insulator-conductor transition of these excitations can take place thereby producing magnetization plateaus.

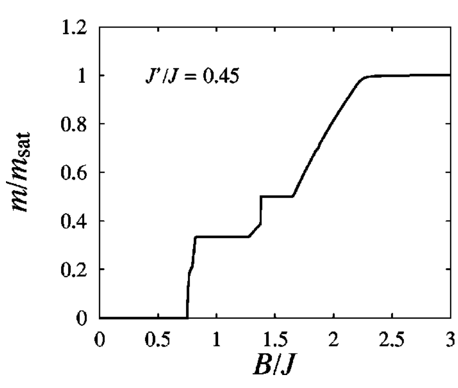

FIG. 5. Magnetization process of the Shastry-Sutherland model with $J'/J=0.45$.

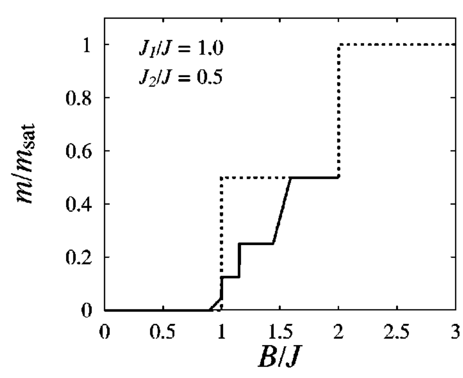

FIG. 7. Magnetization process up to $m/m_{\mathrm{sat}}=0.5$ in the 1/5-depleted square lattice with $J_1/J=1$ and $J_2/J=0.5$. The dotted line shows the case $J_1=J_2=0$.

An example of the plaquette singlet ground states is seen in the $S=1/2$ HAF on the 1/5-depleted square lattice,[11] which includes a possible model for CaV_4O_9 as a special case. The lattice is shown in Fig. 2. In the isolated plaquette limit $J_1=J_2=0$, a trivial plateau already appears at $m/m_{sat}=1/2$ for $J<B<2J$ (see Fig. 7), where every plaquette is in the triplet excited state with $S^z=1$.[16] When $m/m_{sat}<1/2$, the triplet excitations (particle) tend to hop if J_1 and J_2 are turned on, and at specific (commensurate) values of m/m_{sat} they can show insulator-conductor transitions as a consequence of the competition between the hopping and the repulsive interaction. Above $m/m_{sat}=1/2$, a plaquette quintuplet $(S=2)$ with $S^z=2$ behaves as a particle and can show magnetization plateaus between $1/2<m/m_{sat}<1$. In the following, we focus on a weak magnetic-field region, which corresponds to the magnetization $0<m/m_{sat}<1/2$. Regarding the plaquette triplet excitation with $S^z=1$ as a particle and the singlet state as a vacancy, we derive the effective Hamiltonian of the particle by the second-order perturbation around the limit $J_1=J_2=0$. (The explicit form will be shown elsewhere.[14]) The repulsive interactions range from a plaquette to its nearest- and next-nearest neighbors. If parameters satisfy $J_1\simeq 2J_2$, the hopping term is weak and hence the system is in the strong-coupling regime. Then we may expect that the triplets crystallizes and the magnetization plateaus appear at $m/m_{sat}=1/8$ and 1/4. The mean-field approximation of the effective Hamiltonian indeed shows magnetization plateaus at $m/m_{sat}=1/8$ and 1/4 (see Fig. 7). They come from the insulating phases with CDW long-range order of a square structure. A remark is in order here about the relevance of our results to CaV_4O_9. Quite recently, it was shown[17] that plaquettes of another type (*metaplaquettes*) consisting of J_2 bonds play the main role in CaV_4O_9 contrary to earlier studies.[11] The physics is, however, almost the same also in this case; the metaplaquette excitations behave like particles and, J and J_1 bonds induce hopping, and so on. The detailed results will be reported in a longer paper.[14]

Common features. To conclude this paper, we discuss a few features shared by the three examples. According to an analogy to many-particle theories, a plateau state corresponds to a CDW insulating state and gapless ones to supersolids. As the plateau state collapses by increasing the applied field, superfluidity appears, whereas CDW exists in both phases. Let us consider the case of second-order transition, where the magnetization changes continuously. Assuming that the onset of superfluidity is well described by the effective Hamiltonian of the ordinary bosons with a short-range repulsion for low energies, we conclude this transition is of the dynamical exponent $z=2$;[18] magnetization increases linearly like $|H-H_c|$ apart from possible logarithmic corrections. (Note that the form is quite different from that in 1D.)

Also, K. Onizuka *et al.*[19] recently observed a clear 1/3 plateau in $SrCu_2(BO_3)_2$, which we had predicted in this paper and had argued to be of a stripe structure.

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